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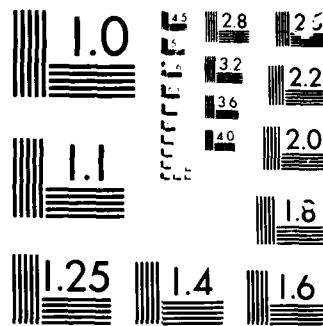UNCLASSIFIED   N00014-79-C-0441   F/G 7/4   NL

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ABSTRACT

The specific heat critical exponent α has been measured for the Au(110) (1x2) order-disorder phase transition using partially integrated Low-Energy Electron Diffraction (LEED) intensities. The resulting value, $\alpha \approx 0.02 \pm 0.05$, is consistent with the predicted Ising universality class of this transition. Evidence for a reduction in the effective critical temperature due to finite size effects is also presented.

Studies of phase transitions on real surfaces provide important tests of our basic ideas about two dimensional critical phenomena. The Au(110)(1x2) surface is an excellent model for studies of continuous order-disorder transitions. The clean surface is inert to the residual gases normally present in an ultra-high vacuum chamber so that much longer times are available for measurements than is usually the case. The phase transition is known to belong to the Ising universality class and the critical exponents, ß, γ and υ have been measured by Campuzano et al. [1]. Thus, Au(110)(1x2) can be used for more refined studies of critical behavior including the role of finite size effects [2] and the kinetics of ordering [3]. In this report we use Au(110)(1x2) to verify a method proposed by Bartelt, Einstein and Roelofs [4] for extracting the heat capacity critical exponent α from low-energy electron diffraction (LEED) data. We also find evidence for finite size effects in this system. It is also significant that the method of BER significantly expands the ability of LEED to study phase transitions since the required measurements and analysis are simple and multiple scattering is fully accounted for.

BER consider LEED intensities integrated over a small region near the center of the surface Brillouin zone. This integration reduces the sensitivity to long range order so that the singular behavior near a phase transition is that of the average energy E. Thus, the exponent α, defined near the critical temperature T_c by [4]

$$E = E_0 + a_1 t \pm b_\pm |t|^{1-\alpha_\pm} + \cdots \qquad (1)$$

where E_0, a_1, b_+ and b_- are constant coefficients, $t = (T-T_c)/T_c$ is the reduced temperature and + and - refer to $t > 0$ and $t < 0$ respectively. Scaling predicts that $\alpha_+ = \alpha_-$ and the ratio b_+/b_- is a universal quantity. For the Ising universality class, $\alpha_\pm = 0$ which implies $b_+/b_- = 1$. As pointed out by BER, most real LEED instruments have a finite transfer width ω and are only sensitive to correlations over a finite range $L_i \propto \omega$ so that the required average is obtained automatically. Furthermore, since multiple scattering is short ranged [4], its presence in LEED does not hinder the

analysis. BER show explicitly, that as long as the diffracted intensity contains no information about the phase of the order parameter, the measured LEED intensity has the form

$$I(t) = \begin{cases} A_0 - A_1 t + B_- |t|^{1-\alpha_-} + \cdots & t < 0 \\ A_0 - A_1 t - B_+ |t|^{1-\alpha_+} + \cdots & t > 0 . \end{cases} \quad (2)$$

where A_0, A_1, B_+ and B_- are constants. As for Equation (1), $\alpha_+ = \alpha_-$ and $B_+/B_- = b_+/b_-$.

BER used Monte Carlo simulations for $(\sqrt{3}\times\sqrt{3})R30$ and $p(2\times2)$ ordered phases on a triangular lattice to show that, depending on the range of t, integration of the intensity over more than about 0.83% of the Brillouin zone were sufficient for Equation (2) to be valid. Since the integral of the intensity over the entire Brillouin zone is conserved [5], the radius of the integration must also be much smaller than the diameter of the zone.

The largest value of t for which Equation (2) is valid is determined by the instrument and the influence of corrections to scaling terms; the smallest depends on the perfection of the surface lattice. The smallest $|t| = t_{min}$ is obtained when the correlation length ξ becomes comparable to the length scale L of the finite size regions;

$$t_{min} \simeq (L/\xi_0)^{-1/\upsilon} . \quad (3)$$

where ξ_0 is the order of several lattice spacings. A more perfect surface results in a small t_{min}. The largest value $|t| = t_{max}$ occurs when ξ is comparable to the transfer width of the instrument, ω;

$$t_{max} \simeq (\omega/\xi_0)^{-1/\upsilon} . \quad (4)$$

unless there are important corrections to scaling beyond those indicated in Eq. (1).

The Au(110) sample is the same one used in a Rutherford Backscattering study [6]. It was oriented to within ±0.5° of the (110) plane and cleaned in ultra-high vacuum with 1 keV argon ion sputtering followed by annealing at 1070 K. Temperature was measured with two type K thermocouples [7] held against the side of the crystal by Pt mounting wires. The temperature was maintained constant to within ±0.2 K during each measurement. Diffracted intensities were measured with a movable Faraday collector whose circular aperture subtended 3.0×10^{-5} sterradians at the sample. A retarding grid behind the aperture was biased 0.9V below the primary beam voltage.

Figure One shows the intensity of the $(0,-\frac{1}{2})$ beam as a function of temperature for a 62 eV primary beam incident at 45° along the $[1\bar{1}0]$ direction. The inflection point provides an estimate of $T_c \simeq 695K$ [4]. Below 650K, the measured full-width at half-maximum (FWHM) of this beam was 0.43 ± 0.02 nm^{-1} in good agreement with the value 0.42 nm^{-1} estimated for our LEED instrument using the procedure described by Park et al. [8]. For the results presented in Figures 1 and 2, this instrument response corresponds to integration over a range of scattered wave vectors equal to about 5.6% of the (1x2) Brillouin zone length along the $[1\bar{1}0]$ direction. Ratios of the FWHM measured for the $(0,-\frac{1}{2})$ and $(0,-1)$ beams were also in good agreement with calculated values and provide evidence that the beam width was dominated by instrumental effects rather than finite size effects. This was not the case for Ref. 1.

Figure One also shows examples of a $(0,-\frac{1}{2})$ beam angular profile measured at 478 K by fixing the angle of incidence and the detector position and varying the incidence energy. Possible artifacts associated with this method are described elsewhere /9/ but are not important for the analysis reported here. The intensity far away from the maximum was used to define a linear background intensity due to thermal diffuse scattering (TDS) and intrinsic surface imperfections. Profiles measured at temperatures far enough above T_c so that fluctuations in the (1x2) phase no longer contribute significantly, show this linear approximation to be accurate to better than 10% of the background intensity. After background subtraction, the

exponential Debye-Waller dependence of the intensity was determined using data with T < 600 K. The effective Debye temperature was found to be 102.5 K in good agreement with previous estimates [6,10]. This Debye temperature dependence was assumed to apply over the entire temperature range of the data, $T \leq 900$ K.

We used the following procedure to analyze the critical behavior of the corrected data. T_c was systematically varied between 640 K and 740 K. For each choice of T_c, A_0 was chosen by a linear extrapolation between the nearest data points on each side of T_c. The data were then plotted on a $\ln(I(t)-A_0)$ vs $\ln|t|$ graph and the longest straight line portion on each side of T_c was determined by a linear least-squares fitting. Examples for T_c values of 680 K, 695 K, and 710 K are shown in Figure Two. The best overall fit was taken to be that which most closely yielded $\alpha_+ = \alpha_-$ and had nearly equal correlation coefficients for $t > 0$ and $t < 0$.

The best fits were obtained for 692 K $< T_c <$ 698 K and the average values of α_+ and α_- over this range of T_c was $\alpha = 0.02 \pm 0.05$. This result is in excellent agreement with the expected Ising value, $\alpha = 0$.

We also estimated the critical exponent $\nu = 1.1 \pm 0.1$ from the measured increase in the FWHM of the $(0,-\frac{1}{2})$ beam in the range 0.02 $< t <$ 0.07. This result agrees with the Ising value, $\nu = 1$ and gives added confidence in our results for α and T_c.

Good fits for α were obtained for

$$0.004 \lesssim t \lesssim 0.035.$$

The diameter of finite size regions, as estimated from Equation (3), is therefore about 250 Au(110) lattice spacings along $[1\bar{1}0]$ or about 100 nm. This result indicates that the substrate was of very good quality and that its contribution to the measured beam FWHM is negligible compared to the instrumental broadening. The value of α is not very sensitive to t_{max} as also pointed out by BER.

We note that the analysis described above has in effect neglected the linear term $A_1 t$ in Equation (2). This is necessitated by the fact that α is zero in the Ising case so that the critical singularity is also linear in $|t|$ to lowest order. The results of BER suggest that this approximation causes the value of α extracted from the analysis to

be too large. Although our analysis does not enable the coefficients $B_\pm$ to be extracted inspection of Figure 2 shows that $(A_1 + B_+)/A_1 + B_-)$ is nearly unity. A more sophisticated fitting procedure is not warranted given the accuracy of the data.

The critical temperature of 695 ± 3 K is substantially larger than the 650 ± 1.5 K obtained by Campuzano et al. [1]. This difference is consistent with that expected from finite size effects. The effective (or pseudo) critical temperature T_e is expected to vary with the scale L of finite size regions approximately as [11]

$$T_c - T_e = \Delta T \simeq aL^{-\nu} T_c.$$

where a is a constant and L is measured in lattice spacings (0.408 nm along the $[1\bar{1}0]$ direction for Au(110)).

Landau [12] has determined a for the case of a finite simple Ising model with free boundary conditions using Monte Carlo simulations and finds $a = 1.25 \pm 0.04$ verifying an earlier result of Ferdinand and Fisher [11] who also present general arguments that a should be of order unity independent of the details of the Hamiltonians. Assuming that our measured T_c is close to that for an ideal surface and taking $T_e = 650$ K from Campuzano et al. [1] yields $L \simeq 20$ lattice spacings. This is in excellent agreement with the mean-size estimated for ordered (1x2) regions on the Au crystal used by Campuzano et al. [1]. They observed the full-width at half-maximum of the fractional order diffraction beam to be about 0.05 of the spacing between integral order beams which also corresponds to ordered regions about 20 substrate lattice spacings wide.

In conclusion, we have measured the specific heat exponent for Au(110)(1x2) using the method of integrated intensities proposed by Bartelt, Einstein and Roelofs [4]. The resulting $\alpha = 0.02 \pm 0.05$ is in excellent agreement with the predicted values and thus verifies the method of integrated intensities. This result also illustrates that the method greatly increases the utility of LEED as a technique to study critical phenomena. We have also presented evidence for a dependence of the effective

critical temperature due to finite size effects consistent with theoretical predictions.
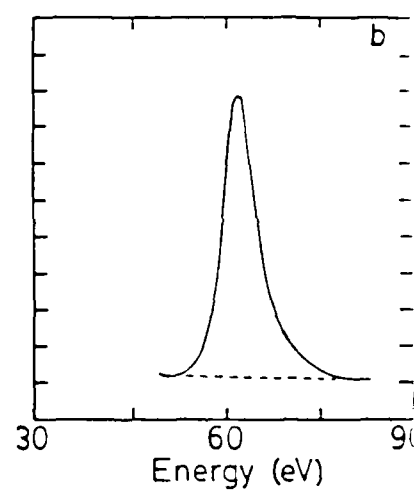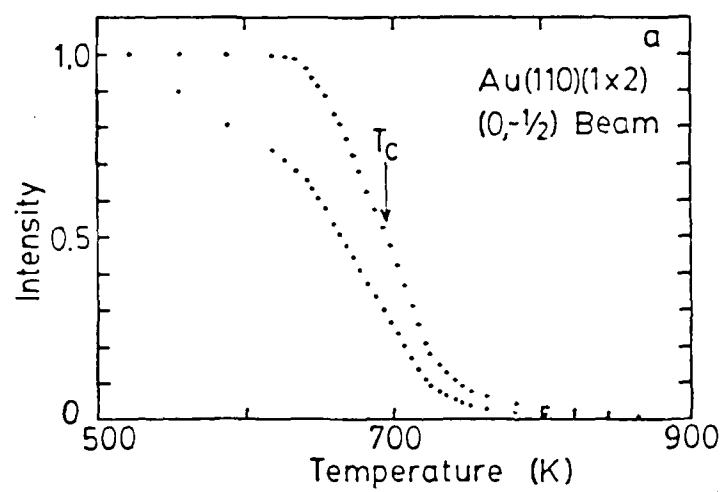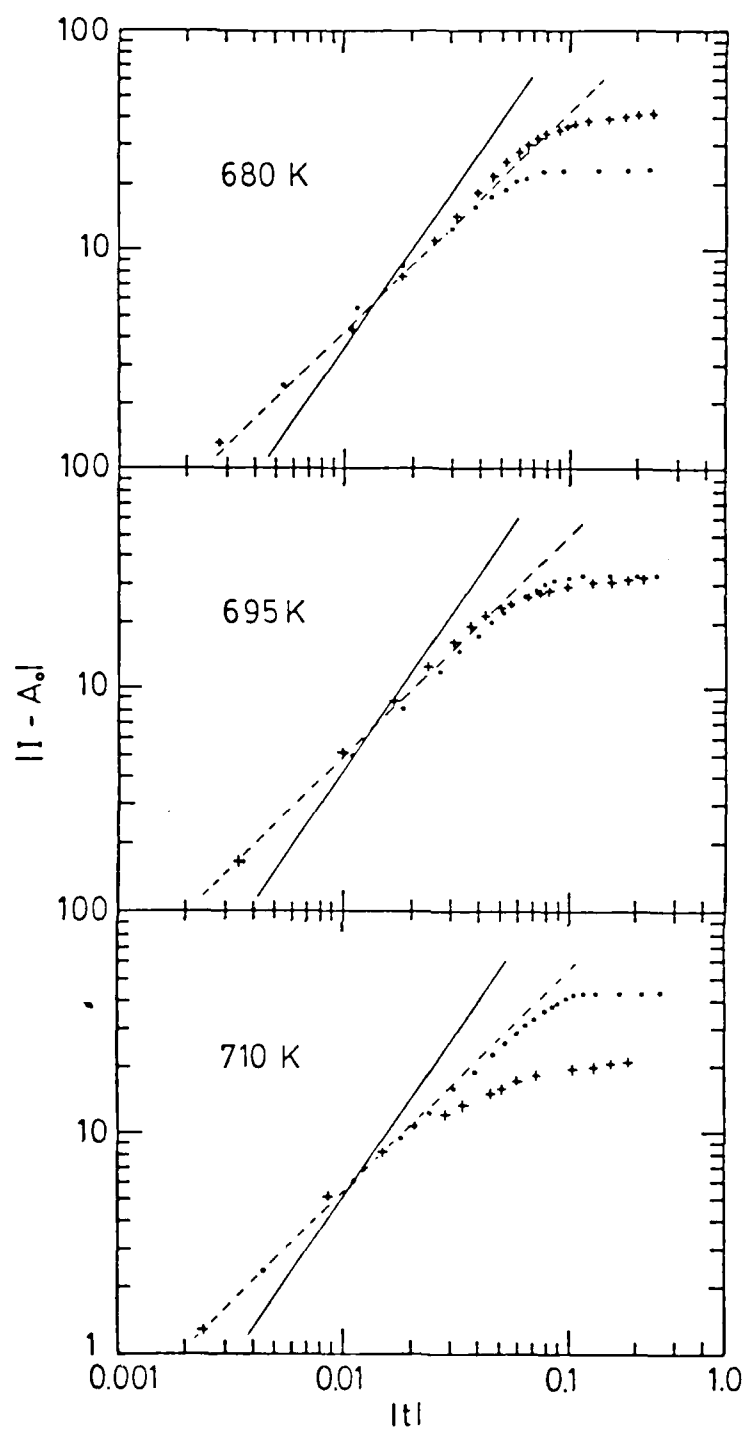
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<u>Figure Captions</u>

Figure One: (a) Measured Intensity vs. Temperature before (o) and after (+) correction for the Debye-Waller factor. (b) $(0,-\frac{1}{2})$ Diffraction beam profile at T – 478 K. The background correction used in the analysis is indicated by the dashed line.

Figure Two: Plots of corrected intensities versus reduced temperature. The dashed lines show the behavior expected for an Ising system with $\alpha = 0$ and the solid lines the behavior for a three state Potts system with $\alpha = 0.33$.

Au(110)(1×2)
(0,-½) Beam

T_c

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